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Improving mechanical strength and thermal shock resistance of SiC/zinc aluminum silicate joint by electrophoretic deposited multi-walled carbon nanotubes

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ABSTRACT

Improving the SiC/zinc aluminum silicate (SiC/ZAS) interface was essential for increasing the mechanical strength of SiC/ZAS joints that were used to join carbon/carbon (C/C) composites and lithium aluminum silicate (LAS) glass ceramics. In this work, well-dispersed multi-walled carbon nanotubes (MWCNTs) were introduced at the SiC/ZAS interface in the joints by electrophoretic deposition, which changed the fracture feature of the bonded C/C-LAS from SiC/ZAS interface fracture to C/C fracture, resulting in significant improvements of the shear strength. Results showed that the average shear strength of the joints was increased by 94.8% after introducing MWCNTs, which was attributed to the obviously decreased micro-defects at the SiC/ZAS interface in the joints. MWCNT-reinforced joints also exhibited better thermal shock resistance evidenced by higher residual shear strengths than those of neat joints after thermal shock tests from 800 °C to room temperature and 1000 °C to room temperature, respectively, which was primarily attributed to the positive role of MWCNTs in alleviating thermal stress.

1. Introduction

Owing to a series of attractive properties including low density, high specific strength and modulus, good anti-friction and anti-wear, excellent thermal shock resistance, low coefficient of thermal expansion (CTE) and so on, carbon/carbon (C/C) composites have been considered as ideal ultra high-temperature structural materials in mechanical engineering, aeronautics and astronautics [1–3]. Lithium aluminum silicate (LAS) glass ceramics possess unique optical properties, low CTE, chemical stability, high-temperature oxidation resistance and thermal shock resistance, which have been widely used in high-temperature electric lighting glass, high-temperature observation window, cookware and large astronomical telescope [4–6]. Joining of these two materials is an attractive strategy to integrate the superior mechanical properties given by C/C composites and the functional features provided by LAS glass ceramics, by which a new generation of multi-functional structural composite materials may be produced.

Recently, the joints of SiC/magnesium aluminum silicate glass ceramic (SiC/MAS) have been adopted on the bonding of C/C composites to themselves or other materials, in which MAS as an interlayer strengthens the joining [7,8] and SiC as a modified layer improves the wettability between C/C composite and glass ceramic [9,10]. Although promising bonding strength has been obtained, the failure of the joints always occurs at the interface between SiC modified layer and glass ceramic interlayer [11,12], suggesting that the interface in the joints is still a weak region needs to be enhanced.

Carbon nanotubes (CNTs) possess small diameters, high specific surface area, low CTE and intrinsically superior mechanical, thermal and electrical performance, making them attractive for enhancing and toughing a variety of matrices and interfaces [13–17]. Moreover, CNTs have been proposed as reinforcing phase of SiC coatings for improving the thermal shock resistance and oxidation resistance of composites [18–20]. Motivated by these researches, providing that CNTs were incorporated into the SiC/glass ceramic joints, it will reinforce the mutual interaction between SiC modified layer and glass ceramic interlayer and meanwhile offer further benefits, i.e., improving the high-temperature resistance of joints capable of being used over a wide temperature range.

In this work, such a joint of SiC/zinc aluminum silicate glass ceramic (SiC/ZAS) reinforced with multi-walled CNTs (MWCNTs) was prepared to join C/C composites to LAS glass ceramics. The first stage involved formation of SiC modified layer on the surface of C/C composites by pack cementation method. The second stage proceeded with the deposition of MWCNTs on the surface of SiC modified layer by electrophoretic deposition (EPD) method. The interest in the EPD technique is driven not only by its applicability to a great variety of materials (e.g., ceramics, coatings and composite materials), but also its high-level efficient process for production of coatings or films from colloidal suspensions at room temperature (RT) [21]. In the last step, the C/C specimens coated with SiC and MWCNT layers were joined to LAS glass ceramics using ZAS as interlayer by vacuum hot-pressing sintering method. Effects of

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the EPD MWCNTs on the microstructure, mechanical strength and thermal shock resistance of the SiC/ZAS joints were investigated.

2. Experimental

2.1. Preparation of SiC modified layer on C/C composites

The C/C specimens (size: 30 mm × 30 mm × 3 mm) with a density of 1.73 g/cm^3 were hand-polished by 200 grit SiC paper, then cleaned in ethanol with an ultrasound bath and dried at 80 °C for 1 h. The powders for preparing SiC layer by pack cementation method were composed of 60–80 wt.% Si, 15–25 wt.% graphite and 5–15 wt.% Al_2O_3. The SiC layer was formed by putting the C/C specimen and as-prepared powders in a graphite crucible and being heat-hearted at 2000–2200 °C for 1–3 h in argon.

2.2. EPD of MWCNTs on the SiC-coated C/C composites

The MWCNT aqueous suspension (purity 3 wt.%) used for EPD deposition was diluted 15-fold with isopropanol and then homogenized by an ultrasound bath for 2 h. During the EPD process, a SiC-coated C/C specimen was fixed on a plastic frame by conductive adhesive as cathode, and then was vertically immersed in the EPD cell containing diluted MWCNT suspension. A stainless steel plate as anode was placed 10 mm away from cathode and the constant voltage was set to be 40 V. SiC-coated C/C specimens deposited with different loadings of MWCNTs were produced by controlling the EPD time (20 s, 40 s and 60 s).

2.3. Joining of C/C composites to LAS glass ceramics

To prepare ZAS raw powders, 15–25 wt.% ZnO, 10–20 wt.% Al_2O_3, 60–75 wt.% SiO_2, 2–5 wt.% TiO_2 and 0.5–1.5 wt.% Sb_2O_3 were thoroughly mixed. Subsequently, the mixture was melted in a corundum crucible at 1500–1600 °C for 2 h, and then quenched to RT in water. Finally, the glass was crushed and ground into powders with the size of 300 meshes. LAS raw powders containing 10–18 wt.% Li_2CO_3, 10–20 wt.% Al_2O_3, 55–75 wt.% SiO_2, 0.5–1% TiO_2, 0.5–1 wt.% B_2O_3 and 0.5–1 wt.% Sb_2O_3 were made in the same way.

Vacuum hot-pressing sintering method was applied to join the C/C specimens coated with SiC and MWCNT layers to LAS glass ceramics using ZAS as the interlayer at the temperature of 1200 °C under pressure of 20 MPa for 45 min. Detailed procedure was described in Ref. [22]. For comparison, the SiC-coated C/C specimens without MWCNT layers were also joined to LAS glass ceramics using ZAS interlayer, i.e., the joint only contained SiC modified layer and ZAS interlayer. The preparation process of the bonded C/C-LAS with MWCNT-reinforced SiC/ZAS joint is depicted in Fig. 1.

2.4. Test and characterization

The test joint specimens were machined into the size of 12 mm × 8 mm × 8 mm. Shear strength of these specimens was measured using a single-lap compressive shear test on a CMT5304-30 kN universal testing machine with a constant loading rate of 0.5 mm/min. Schematic setup for the shear strength tests of the joint specimens is shown in Fig. 2. Five specimens were used for each test.

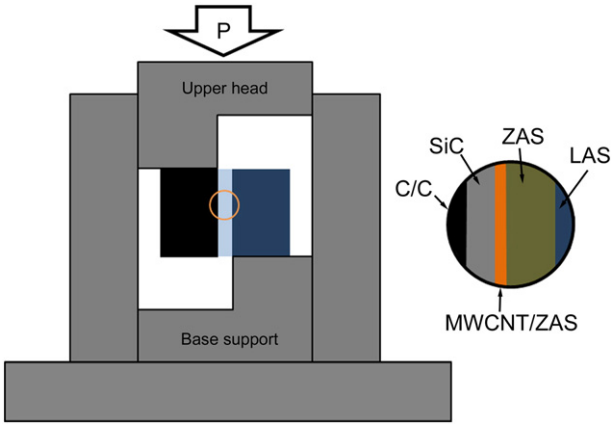

Fig. 2. Schematic setup for the shear tests of the joint specimen.

Thermal shock tests were conducted to investigate the thermal shock resistance of joints. The thermal shock test was carried out by burying the joint specimens in graphite powders to prevent the oxidation of C/C composites. The specimens were put in a furnace with the temperature of 800 °C for 5 min, then pulled out from the furnace directly and kept at RT for 5 min. And then the specimens were put into the furnace again for the next thermal cycle. Thermal shock test between 1000 °C and RT were also performed with the same procedures. Shear strength of the joints was measured after different thermal cycles.

Crystalline phase of the as-prepared SiC modified layer was identified by X-Ray diffraction (XRD, CuKα, X'Pert PRO MPD). Morphological features of the joints were examined by scanning electron microscopy (SEM, JSM-6460) operated at 15 kV, equipped with energy dispersive spectroscopy (EDS). Microstructure of the EPD MWCNTs was investigated by transmission electron microscopy (TEM, Tecnai F30G^2) operated at 200 kV.

3. Results and discussion

3.1. Microstructure of the EPD MWCNTs on SiC modified layer

Fig. 3a is a representative surface SEM image of the as-prepared SiC modified layer coated on C/C composites, showing that the SiC layer has a rough structure composed mainly of big particles. Fig. 3b shows the cross-section backscattering electron (BSE) image of SiC-coated C/C specimens. As seen, the SiC layer has a thickness ranging from 50 μm to 150 μm and bonds with C/C substrate well with no micro-cracks or defects at the interface region. The other observation from Fig. 3b is that SiC has infiltrated to the inside of C/C substrate through open holes in the composites, which is favorable to enhance the interface bonding between SiC modified layer and C/C composite via "nail effect" [7]. XRD pattern (Fig. 3c) reveals that the SiC layer consists of α-SiC, β-SiC and Si phase. After the EPD process, the surface of SiC modified layer is homogeneously covered with a porous MWCNT layer composed of randomly oriented nanotubes, as shown in Fig. 4a–c. It should be noted that the diameter of the pores among MWCNTs decreases with the increase of EPD time. In Fig. 4a, the diameter of pores ranges from 80 nm to 300 nm and the corresponding EPD time is 20 s. In Fig. 4b,

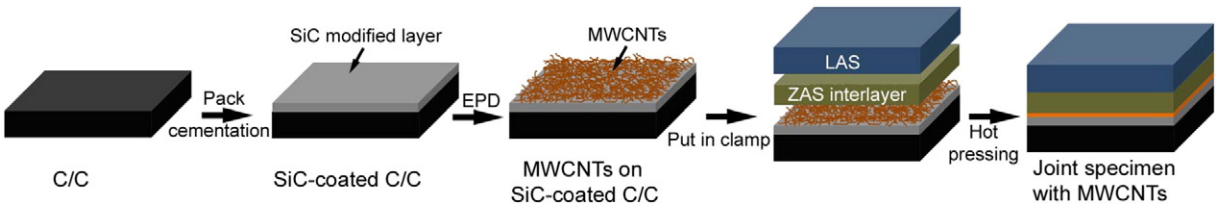

Fig. 1. Schematic of preparation of bonded C/C-LAS with MWCNT-reinforced SiC/ZAS joint.